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Appendix G.3

Phase 2 HGB Mid Course Review

Draft Project H-12 Results: Impact of Biogenic Emissions on Ozone Concentrations in Southeast Texas, August and September 2000

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INTRODUCTION

Biogenic volatile organic compounds (BVOCs), including isoprene (C_5H_8), monoterpenes ($C_{10}H_{16}$), and oxygenated compounds, are emitted in substantial quantities by vegetation (Guenther *et al.*, 2000, Wiedinmyer *et al.*, 2000; Helmig *et al.*, 1999; Kempf *et al.*, 1996; Guenther *et al.*, 1995; König *et al.*, 1995; Fehsenfeld *et al.*, 1992; Winer *et al.*, 1992). The United States Environmental Protection Agency (USEPA) has estimated that total emissions of biogenic VOC emissions in the United States are 30 TgC yr^{-1} , which is an amount greater than estimated anthropogenic VOC emissions. Emissions of isoprene, a highly reactive BVOC, accounted for 17-20 TgC yr^{-1} of the total BVOC inventory (Guenther *et al.*, 2000).

In heavily forested regions of the eastern half of Texas, biogenic emissions of isoprene dominate the emission inventory for reactive hydrocarbons. On a typical summer day, biogenic emissions (primarily isoprene) can exceed 10,000 metric tons day^{-1} (approximately 10 gigagrams day^{-1}) (Wiedinmyer *et al.*, 2001), summed over the eastern half of Texas. In contrast, total anthropogenic emissions of hydrocarbons, summed over the same area, is of the order 2,000 metric tons day^{-1} . While biogenic emissions of hydrocarbons dominate the overall emission inventory in eastern Texas, the spatial distribution of emissions is heterogeneous. In heavily forested areas of eastern Texas, biogenic emissions overwhelm anthropogenic emissions. In highly urbanized areas, anthropogenic emissions are more significant. A number of transition zones exist, where both biogenic and anthropogenic emissions are significant fractions of the emission inventory and the relative roles of biogenic and anthropogenic emissions in ozone formation and other photochemical processes depends on meteorological conditions. Portions of southeast Texas provide examples of this type of transition zone, and in these transition regions, accurately characterizing the spatial distribution of biogenic emissions, over a large spatial domain, is an important element in developing air quality improvement plans.

Developing accurate estimates of the spatial distribution of biogenic emissions relies on accurate characterizations of land covers (leaf biomass densities by species). Wiedinmyer *et al.*

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(2001) has reported on the development of a land use/land cover database for Texas at a spatial scale of 1 kilometer. More recently, the University of Houston (2004) has developed a land use/land cover database for an 8-county region in eastern Texas area at a spatial scale of 4 kilometers. This work will report biogenic emissions, estimated based on these landcover characterizations, during an August-September 2000 photochemical episode. The accuracy of the biogenic emission estimates will be assessed by comparing isoprene concentrations, observed in aircraft measurements, to isoprene concentrations predicted with a photochemical model.

METHODOLOGY

Biogenic emission inventory

The Global Biogenic Emissions and Interactions System (GloBEIS) was used to develop the emission inventory (Yarwood *et al.*, 1999a, b). GloBEIS requires data on land use/land cover (LULC), temperature, leaf area index (LAI), drought index and photosynthetically active radiation (PAR) to estimate biogenic emissions. All the simulations reported in this work used the GloBEIS 3.1 model.

The LULC input data required by GloBEIS 3.1 were derived from two different sources. One source of data, which is referred to as ‘UT/ENVIRON land use data’, was developed by Wiedinmyer *et al.* (2000, 2001). The data are available at a 1-km resolution for a domain encompassing most of Texas. The LULC database contains emission factor data for 156 different vegetation types, including 41 species (e.g., *Quercus alba*), 80 genera (e.g., *Quercus*), and 35 land cover types (e.g., Pecan Elm forest). Each classification is assigned a vegetation species, leaf biomass, and density distribution (Wiedinmyer *et al.*, 2001). In this study, a 4-km resolution database was used. The second source of data is the Texas Forest Service (TFS) data of Byun (2004). The data are available at a 4-km resolution for a domain encompassing the 8-county in Houston-Galveston area; The 8 counties are Galveston, Brazoria, Fort Bend, Waller,

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Harris, Montgomery, Liberty, and Chambers County. This database is referred to as ‘TFS’s land use data’.

Hourly ambient surface temperatures were developed by spatially interpolating temperatures measured by National Weather Service (NWS) and other weather stations throughout southeast Texas (Vizuite *et al.*, 2002). Estimates of PAR flux were based on calculations done by the University of Maryland and the National Oceanic and Atmospheric Administration (NOAA) for the Global Energy and Water Cycle Experiment (GEWEX) Continent Scale International Project (GCIP). NOAA uses a modified version of the GEWEX surface radiation budget (SRB) algorithm (version 1.1) to calculate radiation flux fields from Geostationary Operational Environmental Satellite (GOES-8) data (TCEQ, 2003).

Wind speed and humidity estimates were derived from simulations using the MM5 meteorological model. MM5 is the fifth generation NCAR/Penn State Mesoscale Model. Variable leaf area index (LAI) values are based on a standard 8-day average LAI product from the Moderate Resolution Imaging Spectroradiometer (MODIS). LAI is derived from the atmosphere corrected surface reflectance of two bands (648 and 858 nm) and a 1 km resolution land cover map of six major biomes, characterized by the horizontal and vertical dimensions, canopy height, leaf type, soil brightness and climate, produced from MODIS. LAI, the area of leaves per unit of ground area, is used to determine the amount of leaf biomass and/or model effects due to leaf age in GloBEIS 3.1.

Palmer drought index (PDI), which is an index of moisture deficiency or excess, is empirically derived from the monthly temperature and precipitation scenarios of 13 instances of extreme drought in western Kansas and central Iowa and by assigning an index value of -4 for these cases. Conversely, a +4 represents extremely moist conditions. From these values, categories of wet and dry conditions are defined. The average drought index for eastern Texas in August 2000 was -3.5, which indicates severe drought conditions (http://www.cpc.ncep.noaa.gov/products/analysis_monitoring/regional_monitoring/palmer/2000/08-26-2000.gif). GloBEIS 3.1 assumes that isoprene emissions are not directly influenced by

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drought for a PDI above -2 and that emissions decrease linearly to 10 percent of no-drought conditions for a PDI of -4.

Predictions of isoprene concentrations

Concentrations of isoprene, based on the emissions estimated using GloBEIS, were predicted using the Comprehensive Air Quality Model, with extensions (CAMx, www.camx.com). This model is used by the State of Texas in developing air quality management plans; the State is using CAMx to describe a photochemical episode that occurred in southeast Texas from August 25, 2000 to September 1, 2000. This episode occurred during a large air quality field study and the performance of the model in predicting ozone concentrations has been evaluated using the extensive dataset available from the field program.

CAMx and similar eulerian photochemical grid models simulate emission, advection, dispersion, chemical transformation and physical removal of air pollutants in the framework of a 3-dimensional grid. The horizontal and vertical grid structure used in this work is shown in Figure 1. Wind field inputs were estimated by the Texas Commission on Environmental Quality (TCEQ) using the MM5 meteorological model; emissions (other than biogenic emissions) were based on the emission inventories assembled by the State with some adjustments to the point source data based on ambient observations.

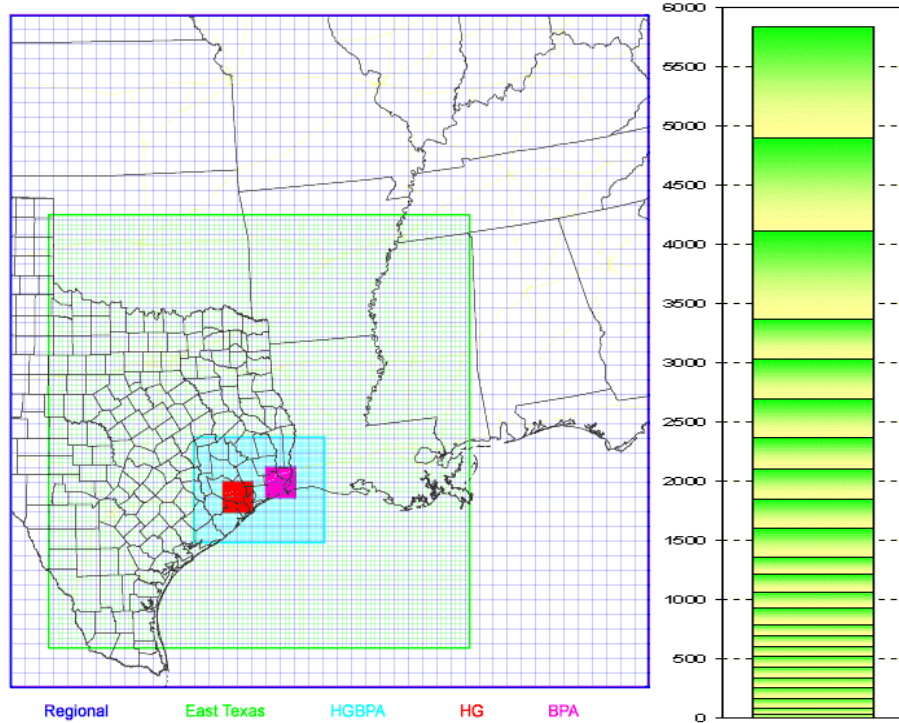


Figure 1. Air quality modeling domain: The domain's horizontal structure consists a coarse grid regional domain (36 km by 36 km resolution) and three nested fine grid subdomains; an East Texas subdomain (12 km by 12 km), Houston/Galveston-Beaumont/Port Arthur subdomain (4 km by 4 km), Houston/Galveston Bay subdomain (1 km by 1 km), and the Beaumont/Port Arthur subdomain (1 km by 1 km). The domain's vertical structure is given on the right hand side of the Figure. Each cell represents a modeling layer and the starting and ending heights of the cells are given in meters above ground level.

Dry deposition model

Dry deposition is the most important physical removal of air pollutants during mid-summer season in Texas (McDonald-Buller et al., 2001). Dry deposition in CAMx is the process determining the resistances with three mechanisms; aerodynamic transport, diffusion across a quasi-laminar sub-layer, and surface uptake. Dry deposition is estimated by the dry deposition rates using two different land use data.

The dry deposition flux is calculated as:

$$F_c = -V_d C_z$$

Where F_c is the dry deposition flux (ppb cm s⁻¹), V_d is the dry deposition velocity (cm s⁻¹), and C_z is the concentration (ppb) at a reference height, usually 10 meters above ground level.

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The dry deposition velocity is calculated as the inverse of the sum of the resistances to deposition:

$$V_d = (R_a + R_b + R_c)^{-1}$$

Where R_a is the aerodynamic resistance between a specified height and the surface ($s\ cm^{-1}$), R_b is the quasi-laminar sub-layer resistance ($s\ cm^{-1}$), and R_c is the bulk surface resistance ($s\ cm^{-1}$) (Wesely et al., 1989).

Application of land use data for estimating dry deposition velocities

Eleven LULC categories are used to estimate dry deposition velocities in CAMx: (1) urban land, (2) agriculture, (3) range, (4) deciduous forest, (5) coniferous forest, (6) mixed forest including wetland, (7) water, (8) barren land, (9) non-forested wetland, (10) mixed agriculture and range land, and (11) rocky open-shrub. Areal fractions of the eleven LULC categories were assigned in each grid cell. Dry deposition velocities for LULC categories were areally weighted to obtain an average dry deposition velocity in each grid cell within the lowest vertical layer of the model. To use land use dataset in CAMx, each land cover category of UT and TFS land use data was mapped to one of the eleven land cover categories used by the dry deposition module in CAMx (McDonald-Buller et al., 2001).

The mapping of TFS land cover categories to CAMx land use categories, used to estimate deposition velocities, are shown in Table 1.

Table 1. Mapping TFS categories to CAMx land use categories

	TFS land cover categories	CAMx LULC categories
1	Impervious	Urban Land
2	-	Agriculture
3	Grass Range	Range Land
4	Forest Broadleaf	Deciduous Forest
5	Forest Coniferous	Coniferous Forest

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6	Forest Mixed	Mixed Forest including Wetland
7	Water	Water
8	Barren	Barren Land
9	-	Non-forest Wetland
10	-	Mixed Agriculture and Range Land
11	-	Rocky Open-shrub

Additional details of the model development, and an evaluation of model performance in predicting ozone concentrations, are available at the TCEQ web site: (http://www.tnrc.state.tx.us/air/aqp/airquality_photomod.html#section4).

Isoprene is modeled as an explicit chemical species in the photochemical modeling, which employed Version IV of the Carbon Bond mechanism (CB-IV). Isoprene reactions with OH·, O₃ and NO₃ are included in the mechanism. The rate constants have been optimized based on chamber experiments (Carter *et al.*, 1996). The values used in the model are 1.476×10^5 for the OH radical reaction, 1.9×10^{-2} for O₃ and 996 ppm min⁻¹ for NO₃ all at 298 K.

Measurements of isoprene concentrations

During the Texas Air Quality Study (TexAQS, www.utexas.edu/research/ceer/texaqs), conducted in August and September, 2000, a variety of measurement platforms recorded isoprene concentrations. Both aircraft and ground measurement platforms were used; however, since the focus of this work is on evaluating the spatial distribution of biogenic emissions, the primary source of isoprene concentration data that will be considered will be from aircraft platforms. A team from the National Oceanic and Atmospheric Administration aboard an Electra aircraft provided by the National Center for Atmospheric Research (NOAA/NCAR Electra) and a team from Brookhaven National Laboratory aboard a G-1 aircraft (BNL G-1) made the bulk of the aircraft based isoprene measurements during TexAQS; both collected air samples in canisters for subsequent laboratory analysis. The NOAA/NCAR Electra typically sampled at an altitude of 600-700 m above ground level (AGL) in the late morning to early afternoon. The air samples typically filled the canister within 10 seconds, with the aircraft flying

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at a speed of approximately 100 m s^{-1} . Isoprene data were measured using both flame ionization and mass spectrometric detection. If both methods yielded results that were above the detection limit, of approximately 1 ppt, an average of both results was used. If one method yield a value above detection limit, and the other did not, the value above the detection limit was used (Personal communication, Donna Sueper, March 13, 2003).

A total of 18 G-1 Research flights were conducted between August 19 and September 11 in 2000. The canister samples were taken aboard the BNL G-1 at altitudes between 400 and 600 m AGL, in the morning to early afternoon. The samples were subsequently analyzed by gas chromatography (ftp://aerosol.das.bnl.gov/pub/Houston00/HYDROCARBONS_V1.txt). Sampling times were 10 seconds. For both aircraft, data on wind speed and directions were obtained.

Methods for comparison of aircraft measurements and model predictions

Air samples, collected by aircraft at 400-600 m AGL, are likely to contain isoprene that was emitted by land covers both immediately below the aircraft, and land covers in a broader area. For this work, the land area footprint was represented by identifying the model grid cells that the aircraft transited through during a 2-minute total period before, during, and immediately after sample collection. Typically, an aircraft would fly through up to six-grid cells within 2 minutes of the collection of the air samples. A composite model prediction was obtained by weighting the data for each grid cell by the length of time that the aircraft spent in the grid cell during the 2-minute period selected for analysis.

RESULTS AND DISCUSSION

Table 2, Table 3, Figure 2 and Figure 3 provide summaries of the comparisons between the model predictions and the aircraft data. The observed data were compared to the isoprene concentrations which are estimated based on (1) dry deposition rate using two LULC data and

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(2) GloBEIS-calculated biogenic emissions also with two LULC data. The mean value of isoprene concentration measured by aircraft was 0.35 ppb; the corresponding modeled values averaged 0.22 ppb for UT land use data and 0.17 ppb for TFS land use data. As shown in Table 2 and Table 3, mean normalized bias and mean normalized gross error for the NOAA dataset were -24% and 73% with UT land use data and -51% and 71% with the TFS land use data; For the BNL G-1 dataset, mean normalized bias and mean normalized gross error were 65% and 121% with the UT land use data and 80% and 127% with the TFS land use data. If the analysis is restricted to NOAA datasets with measured concentrations above 0.1 ppb or 0.2 ppb, then the normalized gross errors of dataset with the UT land use data are reduced to 61% and 48%, respectively and those with the TFS land use data are reduced to 60% and 53%, respectively. The normalized gross errors of G-1 dataset with UT land use data are reduced to 42% and 70%, respectively and those with TFS land use data are reduced to 21% and 12%, respectively. The normalized gross errors of dataset with UT land use data are reduced to 77% and 85%, respectively and those with TFS land use data are reduced to 59% and 51%, respectively.

As shown in Figure 2a, out of a total of 51 measurements made by the NOAA Electra with UT land use data, 21 show agreement between modeled and predicted values that were within a factor of two. In Figure 3a, 16 out of a total of 51 measurements made by the NOAA Electra with TFS land use data showed agreement within a factor of two. In Figure 2b and 3b, 25 out of a total of 52 measurements made by the BNL G-1 aircraft with either case showed agreement within a factor of two. The outliers for the NOAA dataset showed a negative model bias (modeled concentrations lower than observed concentrations), however, those for the BNL G-1 dataset showed a positive model bias (modeled concentrations higher than observed concentrations).

For NOAA dataset, the statistics indicate that the dataset with TFS land use data showed more negative bias compared to that with UT land use data, even though both case showed the similar gross errors. For BNL G-1 dataset, the bias and gross errors with all the samples were

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small for the dataset with UT land use data. However, the dataset with TFS land use data showed lower values of the bias and gross errors with the samples above 0.1 and 0.2 ppb.

Table 2a. Statistical summary of the comparison of predicted isoprene concentrations and NOAA aircraft observations (TFS land use data)

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/25	13	0.39	0.24	-43	69
27	15	0.41	0.20	-58	58
28	14	0.30	0.09	-75	75
30	9	0.27	0.13	-12	88
Total	51	0.35	0.17	-51	71

Samples with measured isoprene concentrations above 0.1 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/25	5	0.69	0.58	14	52
27	5	0.87	0.52	-35	35
28	4	0.54	0.24	-50	50
30	3	0.50	0.23	-28	31
Total	17	0.68	0.42	-23	43

Samples with measured isoprene concentrations above 0.2 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/25	2	1.29	1.16	-9	9
27	5	0.87	0.52	-35	35
28	3	0.65	0.28	-51	51
30	1	1.19	0.39	-68	68
Total	11	0.92	0.56	-38	38

$$\text{Mean Normalized Bias (MNB)} = \frac{1}{N} \sum_{i=1}^N \left(\frac{\text{Pred.} - \text{Obs.}}{\text{Obs.}} \right) \cdot 100\%$$

$$\text{Mean Normalized Gross Error (MNGE)} = \frac{1}{N} \sum_{i=1}^N \left(\frac{|\text{Pred.} - \text{Obs.}|}{\text{Obs.}} \right) \cdot 100\%$$

Where N is the number of observations

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Table 2b. Statistical summary of the comparison of predicted isoprene concentrations and BNL G-1 aircraft observations (TFS land use data)

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/26	25	0.39	0.29	32	98
29	27	0.23	0.24	124	154
Total	52	0.30	0.27	80 %	127 %

Samples with measured isoprene concentrations above 0.1 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/26	13	0.52	0.47	0	80
29	18	0.31	0.29	29	72
Total	31	0.40	0.36	17 %	75 %

Samples with measured isoprene concentrations above 0.2 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/26	8	0.59	0.67	35	95
29	7	0.45	0.36	-20	31
Total	15	0.52	0.52	9 %	65 %

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Table 3a. Statistical summary of the comparison of predicted isoprene concentrations and NOAA aircraft observations (UT land use data)

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/25	13	0.39	0.32	-13	76
27	15	0.41	0.26	-3	76
28	14	0.30	0.13	-65	66
30	9	0.27	0.14	-11	75
Total	51	0.35	0.22	-24	73

Samples with measured isoprene concentrations above 0.1 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/25	8	0.56	0.51	31	70
27	9	0.58	0.37	-28	31
28	5	0.56	0.29	-44	46
30	4	0.43	0.23	-17	23
Total	26	0.55	0.38	-11	45

Samples with measured isoprene concentrations above 0.2 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/25	3	1.07	0.99	-8	23
27	5	0.87	0.57	-29	33
28	4	0.61	0.34	-37	40
30	1	1.19	0.40	-67	67
Total	13	0.86	0.58	-30	35

Table 3b. Statistical summary of the comparison of predicted isoprene concentrations and BNL G-1 aircraft observations (UT land use data)

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/26	25	0.39	0.36	36	113
29	27	0.23	0.24	92	129
Total	52	0.30	0.30	65 %	121 %

Samples with measured isoprene concentrations above 0.1 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/26	14	0.52	0.62	28	116
29	17	0.31	0.30	26	75
Total	31	0.40	0.43	27 %	94 %

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Samples with measured isoprene concentrations above 0.2 ppb

Date of sample collection	Number of pairs	Mean		MNB	MNGE
		Observed	Predicted		
8/26	7	0.51	0.90	119	170
29	9	0.45	0.44	-13	51
Total	16	0.48	0.68	45 %	103 %

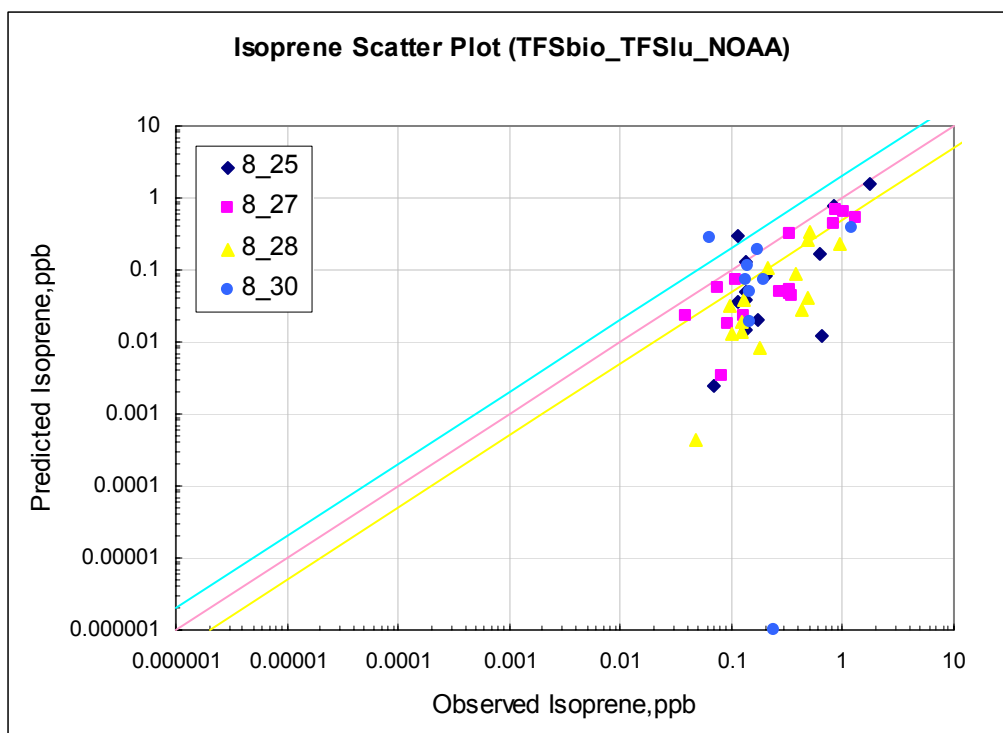


Figure 2a. Scatter plot of predicted and observed isoprene concentrations (NOAA dataset with TFS land use data)

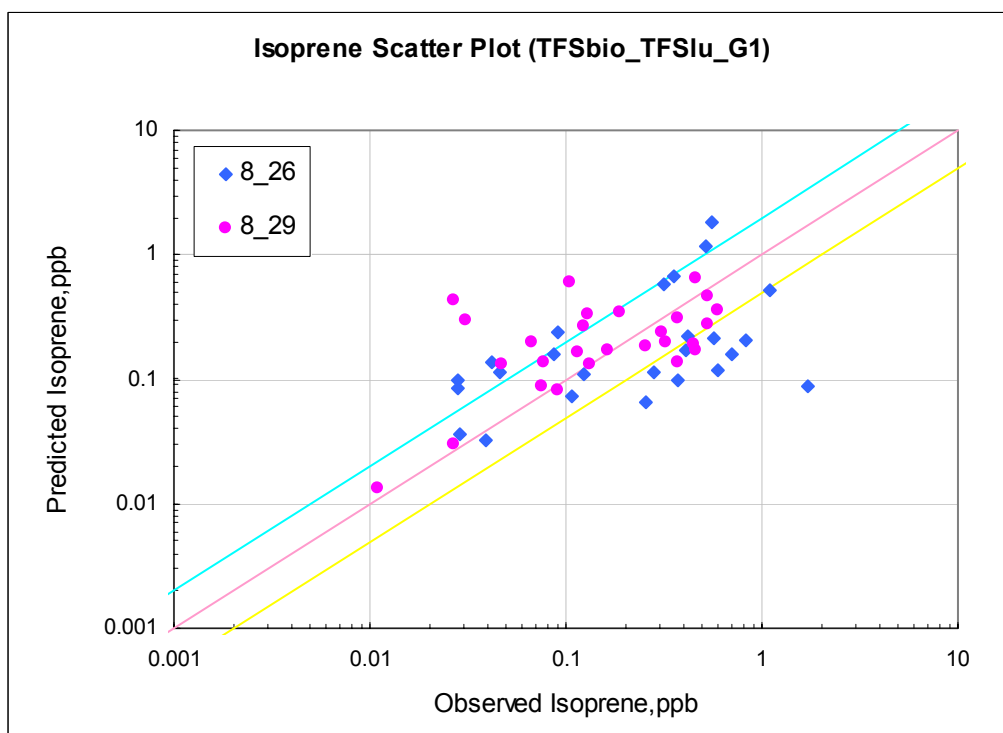


Figure 2b. Scatter plot of predicted and observed isoprene concentrations (BNL G-1 dataset with TFS land use data)

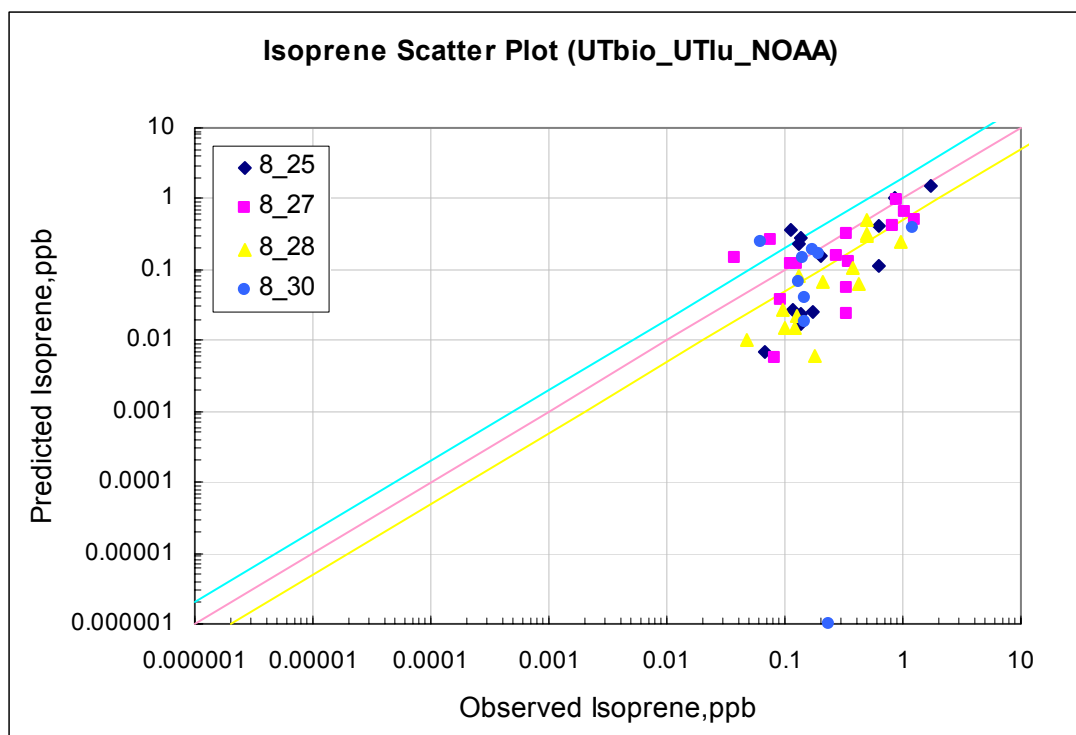


Figure 3a. Scatter plot of predicted and observed isoprene concentrations (NOAA dataset with UT land use data)

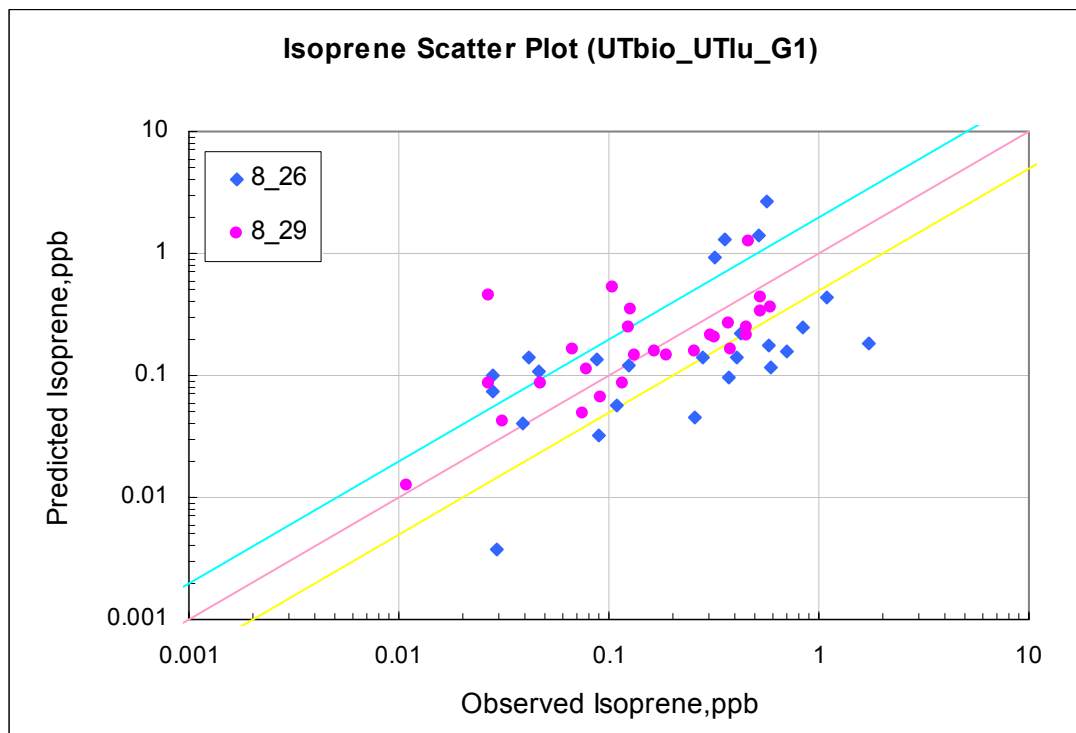


Figure 3b. Scatter plot of predicted and observed isoprene concentrations (BNL G-1 dataset with UT land use data)

CONCLUSIONS

Isoprene concentration measurements made in southeastern Texas during August 2000 were compared to modeled concentrations. For NOAA dataset, normalized gross errors were in the range of 30-80% with a normalized bias of -10 to -50%. For BNL G-1 dataset, normalized gross errors were in the range of 10-80% with a normalized bias of 60-130%. Normalized gross errors tended to decrease at higher observed concentrations. The greatest discrepancies between modeled and predicted concentrations for NOAA dataset were due to model under-predictions of observed concentrations. By contrast, the discrepancies for BNL G-1 dataset were due to model over-prediction of observed concentrations. Between two cases for NOAA dataset, that with UT land use data showed the better performance. For BNL-G1 dataset, that with UT land use data showed the better performance for all samples, however, that with TFS land use data performed better for the samples with the isoprene concentrations above 0.1 and 0.2 ppb.

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